PATENT COOPERATION TREATY

PCT

REC'D 11 JAN 2005
WIPO PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference F17564 AS/vd	**FOR FURTHER ACTION**	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/IB 03/04701	International filing date *(day/month/year)* 23.10.2003	Priority date *(day/month/year)* 25.10.2002
International Patent Classification (IPC) or both national classification and IPC C08J7/06		
Applicant SOUTH AFRICAN NUCLEAR ENERGY CORPORATION LIMITED e		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 6 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 10 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the opinion
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
30.03.2004	10.01.2005
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Müller, M Telephone No. +49 89 2399-8665

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/IB 03/04701

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, Pages

2-6, 8-50	as originally filed
1, 7	received on 09.09.2004 with letter of 08.09.2004

Claims, Numbers

1-38	received on 09.09.2004 with letter of 08.09.2004

Drawings, Sheets

1/2-2/2	as originally filed

2. With regard to the **language,** all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).

   ☐ the language of publication of the international application (under Rule 48.3(b)).

   ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   ☐ contained in the international application in written form.

   ☐ filed together with the international application in computer readable form.

   ☐ furnished subsequently to this Authority in written form.

   ☐ furnished subsequently to this Authority in computer readable form.

   ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   ☐ the description, pages:

   ☐ the claims, Nos.:

   ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	2-35
	No:	Claims	1,36-38
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-38
Industrial applicability (IA)	Yes:	Claims	1-38
	No:	Claims	

2. Citations and explanations

   see separate sheet

re item V

Cited documents

D1: US-A1-2001009176
D2: JP(A) 58199132
D3: GB-A-1488931
D4: US-A-4800053

Novelty (Article 33(2) PCT)

General remarks

(i) Any argument related to HF as fluorine containing gas is irrelevant due to restriction of the fluorine containing gas in claim 1 to gases different from HF.

(ii) Any argument that it is not F_2 that reacts in the prior art with the surface of the solid bonding member but any gas being formed from F_2 is unsubstantiated and as such must be disregarded. Further, it is not excluded in claim 1 that the reaction of the fluorine containing gas occurs via an intermediate. Also for this reason, such an argument is not acceptable. Finally, should the F_2 in D1 not react with said bonding member surface while it does in the claimed invention, it would not be clear which measure has to be applied to arrive at such reaction. The latter would amount to a deficiency under Article 5 PCT.

(iii) Any statement that the oxygen applied in the prior art does not react is unsubstantiated and as such must be disregarded. Further, should the oxygen in the prior art not react while it does in the claimed invention, it would not be clear which measure has to be applied to arrive at such reaction. The latter would amount to a deficiency under Article 5 PCT.

(iv) Any attempt to create a distinguishing feature by stating that the water in the claimed process has to react with the bonding member surface while it does not in the prior art must fail as no such reaction is cited in claim 1 of the present application.

Novelty over the cited documents

D1 (paragraphs [0142] and [0143] and all working examples) discloses a process of contacting a solid bonding member with

(i) HF,
(ii) air and
(iii) water vapour.

In paragraph [0136], it is explicitly disclosed that alternatively to HF, F_2 can be used as **reactive** fluoride gas. Hence D1 discloses a combination of all components cited in

claim 1 of the present application, cites the contacting of a solid bonding member with said combination, and finally discloses that F_2 is a reactive fluoride gas.

D2 (abstract) discloses treating a rubber material with
(i) fluorine gas, and an oxygen generating component comprising
(ii) Fe_2O_3, NiO, CaO or MgO and
(iii) water.
Apart from the above general remarks, it is noted with respect to D2 that this document explicitly discloses a treatment of the rubber material which implies reaction with said material.

D3 (page 6, lines 33 - 38) discloses treating nylon (a polyamide) with
(i) fluorinating gas with the only fluorinating gas disclosed being F_2,
(ii) 1 - 5% oxygen and
(iii) 4% water.
Apart from the above general comments, it is noted with respect to D3 that the question of whether the presence of air or water is disclosed in D3 to be disadvantageous is irrelevant to the question of novelty. The only issue that matters for novelty is whether such presence is disclosed or not. Further, the issue of whether D3 discloses only minor amounts of water is irrelevant to the question of novelty as well as the present claims are not restricted to a particular water or oxygen amount.

D4 (example 1) discloses treating polypropylene with
(i) fluorine and air, i.e. a gaseous mixture comprising
(ii) oxygen and
(iii) water vapour.
Apart from the above general comments, it is noted with respect to D4 that, as acknowledged by the applicant, the Shorter Oxford English dictionary defines air as the gaseous substance which envelopes the earth and is breathed by all land animals and plants. There will be no doubt that the air enveloping the earth contains humidity, i.e. water. The disclosure of air in D4 therefore without any doubt implicitly includes the disclosure of water.

The subject-matter of at least independent claims 1 and 37 as well as claims 36 and 38 thus lacks novelty over any of D1 - D4.

Inventive step (Article 33(3) PCT)
It is not clear which problem is solved by the claimed subject-matter. The latter hence

lacks at least inventive step over the cited references. It is noted in this respect that any advantage derived from the presence of water during oxyfluorination cannot establish inventive step as, as has been shown above, said feature has already been disclosed in any of D1 - D4 and any non-distinguishing feature is irrelevant to the assessment of inventive step.

OXYFLUORINATION

THIS INVENTION relates, broadly, to oxyfluorination. More particularly the invention relates to a process for the oxyfluorination of a surface of a solid to activate it.

According to the invention, there is provided a process for the activation by oxyfluorination of at least part of a surface of a solid, which process includes exposing, under selected conditions of temperature and pressure and for a selected reaction time, at least part of the surface of the material of the solid to an oxyfluorinating atmosphere which is a gas/vapour mixture which includes at least one fluorine-containing gas which reacts with the material of the exposed surface, at least one oxygen-containing gas which reacts with the material of the exposed surface, and water vapour, said gases in the oxyfluorinating atmosphere acting to oxyfluorinate the exposed surface, thereby to activate it, and the water vapour acting to enhance the activation of the exposed surface to enhance the amenability of the exposed surface to adhesive bonding to other materials, the process including selecting the fluorine-containing gas from the group consisting of F_2, XeF_2, ClF, ClF_3, BrF, BrF_3, BrF_5, IF_7, OF_2, O_2F_2 and mixtures of any two or more thereof.

By fluorine-containing gas is meant that each molecule of the gas contains at least one fluorine atom, and the term oxygen-containing gas has a corresponding meaning.

Instead, the process may include selecting the solid material which is subjected to activation by oxyfluorination from metals and metalloids which are members of the group consisting of mild steel, low carbon steel, stainless steel, and mixtures or alloys of any two or more thereof. In particular, the process may thus include selecting mild steel or low carbon steel as the solid material which is subjected to activation by oxyfluorination.

As will be appreciated and as indicated above, exposing the surface of the solid material to the oxyfluorinating atmosphere comprising the gas/vapour mixture of the present invention will be under conditions of temperature and pressure, and for a reaction time, selected to provide the exposed surface with desired properties such as, in particular, an enhanced amenability to adhesive bonding to other materials. In particular, the process may include selecting the fluorine-containing gas which reacts with the exposed surface from the group consisting of molecular fluorine (F_2), fluorinated noble gases, fluorohalogens, oxides of fluorine, and mixtures of any two or more thereof. As indicated above, the fluorine-containing gas may be molecular fluorine (F_2) itself, or it may be made up of one or more other suitable fluorine-containing gaseous compounds, examples of which are fluorinated noble gases such as XeF_2, or fluorohalogens such as ClF, ClF_3, BrF, BrF_3, BrF_5, and IF_7, or oxides of fluorine such as OF_2 or O_2F_2 so that, in other words, the

CLAIMS:

1. A process for the activation by oxyfluorination of at least part of a surface of a solid, which process includes exposing, under selected conditions of temperature and pressure and for a selected reaction time, at least part of the surface of the material of the solid to an oxyfluorinating atmosphere which is a gas/vapour mixture which includes at least one fluorine-containing gas which reacts with the material of the exposed surface, at least one oxygen-containing gas which reacts with the material of the exposed surface, and water vapour, said gases in the oxyfluorinating atmosphere acting to oxyfluorinate the exposed surface, thereby to activate it, and the water vapour acting to enhance the activation of the exposed surface to enhance the amenability of the exposed surface to adhesive bonding to other materials, the process including selecting the fluorine-containing gas from the group consisting of F_2, XeF_2, ClF, ClF_3, BrF, BrF_3, BrF_5, IF_7, OF_2, O_2F_2 and mixtures of any two or more thereof.

2. A process as claimed in Claim 1, in which the enhancement of the activation of the exposed surface acts to enhance the amenability of the exposed surface to adhesive bonding to other materials.

3. A process as claimed in Claim 1 or Claim 2, which includes selecting the solid material which is subjected to activation by fluorination from the group consisting of polymeric materials having constituents which are confined to carbon and hydrogen, elastomeric materials having constituents which are confined to carbon and

hydrogen, polymeric materials having constituents which are not confined to carbon and hydrogen and which include, in addition to carbon and hydrogen, other atomic species as constituents, elastomeric materials having constituents which are not confined to carbon and hydrogen and which include, in addition to carbon and hydrogen, other atomic species as constituents, carbon, glasses, metals, metalloids, wood, leather, cotton, wool, ceramics, asbestos and blends and mixtures thereof.

4. A process as claimed in Claim 3, which includes selecting the solid material which is subjected to activation by oxyfluorination from the group of materials consisting of polymeric materials, elastomeric materials and mixtures of any two or more thereof.

5. A process as claimed in Claim 4, which includes selecting the solid material which is subjected to activation by oxyfluorination from the group of materials having constituents which are confined to carbon and hydrogen.

6. A process as claimed in Claim 4, which includes selecting the solid material which is subjected to activation by oxyfluorination from the group of materials having constituents which are not confined to carbon and hydrogen and which include, in addition to carbon and hydrogen, other atomic species as constituents.

7. A process as claimed in Claim 3, which includes selecting the solid material which is subjected to activation by oxyfluorination from the group of materials

consisting of carbon, glasses, metals, metalloids and mixtures of any two or more thereof.

8. A process as claimed in Claim 7, which includes selecting carbon as the material which is subjected to activation by oxyfluorination.

9. A process as claimed in Claim 7, which includes selecting the solid material which is subjected to activation by oxyfluorination from metals and metalloids which are members of the group consisting of mild steel, low carbon steel, stainless steel and mixtures or alloys of any two or more thereof.

10. A process as claimed in Claim 9, which includes selecting mild steel or low carbon steel as the solid material which is subjected to activation by oxyfluorination.

11. A process as claimed in any one of the preceding claims, which includes selecting the fluorine-containing gas which reacts with the exposed surface from the group consisting of molecular fluorine, fluorinated noble gases, fluorohalogens, oxides of fluorine, and mixtures of any two or more thereof.

12. A process as claimed in any one of the preceding claims, which includes selecting the oxygen-containing gas which reacts with the exposed surface from molecular oxygen, ozone and mixtures thereof.

13. A process as claimed in any one of the preceding claims, which includes diluting the oxyfluorinating atmosphere with a diluent gas which is inert to the exposed surface and inert to the other constituents of the oxyfluorinating atmosphere, and does not react therewith.

14. A process as claimed in Claim 13, which includes selecting the inert gas from the group consisting of nitrogen, the noble gases and mixtures of any two or more thereof.

15. A process as claimed in Claim 14, which includes selecting the inert gas from the group consisting of helium, argon, carbon dioxide, molecular nitrogen and mixtures of any two or more thereof.

16. A process as claimed in any one of the preceding claims, which includes using, as the oxyfluorinating atmosphere, a gas/vapour mixture of molecular fluorine, molecular oxygen and water vapour.

17. A process as claimed in Claim 16, which includes diluting the oxyfluorinating atmosphere, using molecular nitrogen as a diluent.

18. A process as claimed in any one of the preceding claims, which includes subjecting the oxyfluorinating atmosphere to ultra-violet radiation before the exposing of the solid material to the oxyfluorinating atmosphere is ended.

19. A process as claimed in Claim 18, in which the subjecting of the oxyfluorinating atmosphere to ultra-violet radiation is prior to the exposing of the solid material to the oxyfluorinating atmosphere.

20. A process as claimed in Claim 18 or Claim 19, in which the subjecting of the oxyfluorinating atmosphere to ultra-violet radiation is during the exposing of the solid material to the oxyfluorinating atmosphere.

21. A process as claimed in any one of the preceding claims, which includes exposing the solid material to a said oxyfluorinating atmosphere in which the fluorine-containing gas includes molecular fluorine at a partial pressure of 0.01 – 200 kPa.

22. A process as claimed in any one of the preceding claims, which includes exposing the solid material to the oxyfluorinating atmosphere for a period of 0.10 seconds – 10 hours, at a total pressure of the oxyfluorinating atmosphere of 0.1 – 500 kPa with the surface of the solid material and the oxyfluorinating atmosphere at a temperature at which the solid material has a surface which is stable.

23. A process as claimed in Claim 22, which includes exposing the solid material to the oxyfluorinating atmosphere at a total pressure of 1 – 200 kPa, and at a said temperature which is above 0°C, for a period of 0.1 seconds – 1 hour.

24. A process as claimed in Claim 23, which includes exposing the solid material to the oxyfluorinating atmosphere at a pressure 5 – 150 kPa and at a said temperature which is 20 – 100°C, for a period of 1 second – 10 minutes.

25. A process as claimed in any one of the preceding claims, which includes exposing the solid material to a said oxyfluorinating atmosphere which, in addition to its comprising a fluorine-containing gas, an oxygen-containing gas and water vapour, includes at least one further reactive constituent selected from the group consisting of halogens other than fluorine, interhalogen compounds and mixture of any two or more thereof.

26. A process as claimed in any one of the preceding claims, which includes exposing the solid material to a said oxyfluorinating atmosphere having a fluorine-containing gas content of 0.1 – 99% by volume.

27. A process as claimed in Claim 26, which includes exposing the solid material to a said oxyfluorinating atmosphere having a fluorine-containing gas content of 1 – 30% by volume.

28. A process as claimed in Claim 26 or Claim 27, which includes exposing the solid material to a said oxyfluorinating atmosphere in which the fluorine-containing gas forms 5 – 20% by volume and the oxygen-containing gas forms 5 – 95% by volume.

29. A process as claimed in any one of Claim 26 – 28 inclusive, which includes exposing the solid material to a said oxyfluorinating atmosphere which has a relative humidity of 0.1 – 99%.

30. A process as claimed in Claim 29, which includes exposing the solid material to a said oxyfluorinating atmosphere which has a relative humidity of 30-90%.

31. A process as claimed in Claim 29, which includes exposing the solid material to a said oxyfluorinating atmosphere which has a relative humidity of 50 – 80%.

32. A process as claimed in any one of the preceding claims, which includes exposing the solid material to the oxyfluorinating atmosphere until the surface concentration of fluorine of said exposed surface has been increased by at least 0.01 $\mu gF/cm^2$.

33. A process as claimed in Claim 32, which includes exposing the solid material to the oxyfluorinating atmosphere until the surface concentration of fluorine of said exposed surface has been increased by 0.01 – 50 $\mu gF/cm^2$.

34. A process as claimed in any one of the preceding claims, which includes, prior to the exposing of the solid material to the oxyfluorinating atmosphere, degreasing the exposed surface.

35. A process as claimed in any one of the preceding claims, which includes exposing the solid material to the oxyfluorinating atmosphere in a reaction chamber in a reaction vessel, and which includes flushing the reaction chamber by means of the oxyfluorinating atmosphere prior to the exposing of the solid material to the oxyfluorinating atmosphere.

36. A process as claimed in Claim 1, substantially as described herein.

37. An oxyfluorinated product whenever produced by the process of any one of Claims 1 – 37 inclusive.

38. A product as claimed in Claim 38, substantially as described herein.